

Organochlorine and PBDE Concentrations in Relation to Cytochrome P450 Activity in Livers of Forster's Terns (*Sterna forsteri*) and Caspian Terns (*Hydroprogne caspia*), in San Francisco Bay, California

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Abstract We measured halogenated organic contaminants (HOCs) [polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and dichloro-diphenyl-trichloroethane (DDT)] and P450 [e.g., ethoxyresorufin-*O*-dealkylase (EROD)] stress in livers from Caspian tern (*Hydroprogne caspia*) adults and Forster's tern (*Sterna forsteri*) adults and chicks in San Francisco Bay (SFB). Penta BDEs and tetra PBDEs composed 46–66% of \sum PBDE in terns. PCB homologues di, tri, penta, hexa, and hepta composed 93–95% of \sum PCBs and *p,p'*-DDE composed 82–98% of all \sum DDTs. We found similar concentrations of \sum PBDEs [mean micrograms per gram wet weight (ww) \pm standard error = 0.4 ± 0.1], \sum PCBs (5.9 ± 1.6), and \sum DDTs (0.6 ± 0.1) among species, sexes, and regions. However, concentrations were higher in Forster's tern adults than chicks (\sum PBDEs = 0.4 ± 0.1 and 0.1 ± 0.1 ; \sum PCBs = 7.08 ± 2.4 and 2.4 ± 1.4 ; \sum DDTs = 0.5 ± 0.1 and 0.1 ± 0.1 ; respectively), and there was a

nonsignificant trend of elevated \sum PBDEs and \sum PCBs for adult Forster's terns in the Central South Bay and Lower South Bay portions of SFB. Combined Forster's tern and Caspian tern \sum DDTs bioaccumulated similarly to selenium, but not mercury, and there was a nonsignificant but positive trend for \sum PBDEs and \sum PCBs bioaccumulation with mercury. P450 protein activity was higher in adult Forster's terns than Caspian terns, higher in Central South Bay than in Lower South Bay, and higher in adult Forster's terns than in chicks.

San Francisco Bay (SFB) and adjacent wetlands provide critical breeding, stopover, and wintering habitat for waterbirds, with 25% of the Pacific Flyway waterfowl population and up to one million shorebirds using the estuary annually (Page et al. 1999; Stenzel et al. 2002). SFB wetlands also have been designated as a site of hemispheric importance for shorebirds (Harrington and Perry 1995), and ~30% of the Pacific coast breeding population of Forster's terns nest within SFB wetlands (McNicholl et al. 2001; Strong et al. 2004). However, SFB has become highly urbanized over the past century, which has subjected it to extensive pollution from environmental contaminants and has resulted in the loss of up to 90% of SFB wetlands. Together, these anthropogenic impacts have adversely affected the health of the SFB ecosystem (Goals Project 1999).

San Francisco Bay is contaminated by an extensive suite of pollutants (Davis et al. 2003; Davis et al. 2007; Thompson et al. 2007) and is currently listed under section 303(d) of the Federal Clean Water Act as being impaired by polychlorinated biphenyls (PCBs), dioxin and furan compounds, legacy pesticides [dichloro-diphenyl-trichloroethane (DDT)], chlordane, dieldrin, and trace

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elements [mercury (Hg) and selenium (Se)] (San Francisco Bay Regional Water Quality Board 2007). Additionally, recent reports have highlighted elevated levels of polybrominated diphenyl ethers (PBDEs) in SFB (North 2004; Oros et al. 2005). Although there has been a large-scale effort to regularly monitor SFB contaminants in water, sediment, and fish throughout the region (e.g., Davis et al. 2007; Johnson et al. 2000; Oros et al. 2005; She et al. 2000, 2008), the concentrations and impacts of halogenated organic compounds (HOCs) such as PBDEs, PCBs, and DDT on local waterbirds are less well understood (Thompson et al. 2007). The high trophic position of fish-eating waterbirds makes them particularly vulnerable to highly bioaccumulative compounds. Moreover, the significance of the SFB area for waterbird populations highlights the importance of understanding current trends of these HOC contaminants in SFB waterbirds.

In this study, we measured the concentrations of several groups of contaminants of concern in two fish-eating waterbird species that breed within the SFB: Caspian terns (*Hydroprogne caspia*) and Forster's terns (*Sterna forsteri*). Terns are ideal species to use as bioindicators of contaminant exposure in the SFB because they are piscivorous (McNicholl et al. 2001) and thus forage at a higher trophic level than other waterbird species and nest and forage in wetlands throughout the SFB (Ackerman et al. 2008a; Bluso-Demers et al. 2008; Strong et al. 2004). However, these two species also exhibit different foraging ecologies. Caspian terns tend to forage in the open bay (Anderson et al. 2007; Lyons et al. 2005) on larger fish (>10 cm; Cuthbert and Wires 1999), whereas Forster's terns tend to forage in wetlands along the bay's margins (Ackerman et al. 2008a) on smaller fish (<10 cm; McNicholl et al. 2001). Thus, each species provides an indication of contaminant exposure, uptake, and bioaccumulation in different SFB habitats.

Waterbird exposure to contaminants is concerning because it might adversely affect body condition, survival, and reproduction (Hoffman et al. 1998, 2005). Quantifying the effect of contaminants on these end points in field studies can be difficult and costly; thus, biochemical responses are often measured to indicate potential impairment. One common measured response associated with contaminant exposure is cytochrome P450 induction. Elevated levels of P450 proteins [e.g., ethoxyresorufin-*O*-dealkylase (EROD)] reflect the metabolism of toxic compounds (lipophilic xenobiotics) into more water-soluble, excretable forms (Sijm et al. 1993; Stegeman and Hahn 1994).

Our objectives were to (1) establish baseline data on concentrations of HOCs in Caspian tern adults and Forster's tern adults and chicks in the SFB, (2) determine if concentrations of HOCs differed by species, region, sex,

and age (adults and chicks), (3) determine if HOCs bioaccumulated similarly to Hg and Se, and (4) evaluate whether current HOC exposure in the SFB might be inducing a cytochrome P450 response.

Methods

Bird Collection

During 2005, we collected whole birds [$n = 72$; 62 Forster's terns (adults, $n = 46$; chicks, $n = 16$) and 10 Caspian terns] using either a shotgun (with steel shot), bow net, or by hand at three regions within the SFB: North Bay ($n = 31$; Napa Marsh Restoration Area; 38°08'N 122°18'W), Central South Bay ($n = 9$; Eden Landing Ecological Reserve; 37°36'N 122°08'W), and Lower South Bay ($n = 32$; Don Edwards San Francisco Bay National Wildlife Refuge; Pond A1, Pond A11, New Chicago Marsh; 37°26'N, 121°58'W). A detailed description of the sites and a map of the study site can be found in Eagles-Smith et al. (2009b). We sampled breeding Caspian terns ~400 m from their colonies while they were either departing from or returning to colonies (4–21 April). Prebreeding Forster's tern adults were sampled while foraging in wetlands along the Bay's margins (13–21 May), whereas breeding Forster's terns were captured on their nests (26 May–1 June). Forster's tern chicks were captured by hand within colonies during routine nest monitoring (13 July–8 August). Sexes were determined via necropsy and confirmed via DNA analysis (Ackerman et al. 2008c; Bluso et al. 2006; Zoogen 2008). We removed the liver immediately after collection using acid- and hexane-rinsed stainless-steel forceps, scalpels, and scissors. Small aliquots [~ 1 g wet weight (ww)] were subsampled from the tip of the larger lobe of each liver for P450 measurements and placed in sterile, polypropylene cryovials. The remaining liver tissue samples were stored in chemically cleaned I-CHEM glass vials (Chase Scientific Glass, Rockwood, TN, USA). All samples were immediately frozen on dry ice until returned to the laboratory, within 48 h. Samples for P450 measurements were stored at -80°C until analysis, and the remaining livers were stored at -20°C until processing and contaminant analyses. We collected all birds under U.S. Fish and Wildlife Service and California Department of Fish and Game scientific collecting permits as well as the Animal Care and Use Committee, Western Ecological Research Center, U.S. Geological Survey.

PCBs, PBDEs, and Organochlorine Pesticides

Liver tissue samples were analyzed for 101 PCB congeners, 6 legacy pesticide compounds, and 38 PBDE

congeners (Table 1) by the Geochemical and Environmental Research Group, Texas A&M University, College Station, Texas, USA. Briefly, tissue samples analyzed for PCBs and pesticides were extracted following NOAA Status and Trends Method (MacLeod et al. 1985) with minor revisions (Brooks et al. 1989; Wade et al. 1988). Lipid content was first determined using a Soxhlet extraction apparatus (Dobush et al. 1985). Samples (1–10 g ww) were extracted by adding surrogate standards, Na₂SO₄, and methylene chloride in a centrifuge tube. Tissue extracts were purified by silica/alumina column chromatography to isolate the aliphatic and PCB/pesticide fractions. PCB/pesticide fractions were further purified by high-performance liquid chromatography (HPLC) in order to remove interfering lipids. Quantitative analyses were then performed by capillary gas chromatography (CGC) with a flame ionization detector for aliphatic hydrocarbons, CGC with electron capture detector for PCBs and pesticides, and a mass spectrometer detector in the SIM mode for PBDEs (Wade et al. 1988). PCBs and pesticides were initially analyzed on a DB-5 capillary column, and analyte identity and concentrations were confirmed on a DB-17 capillary column. Because of the restrictive costs of extensive HOC laboratory analyses, we only included a random subset of the total birds collected for the HOC analyses [$n = 18$; 13 Forster's terns (adults, $n = 7$; chicks, $n = 6$) and 5 Caspian terns] but used our larger dataset to assess the P450 response.

Mercury and Selenium

Liver total Hg (THg) and Se concentrations from these birds were reported elsewhere (Ackerman and Eagles-Smith 2009; Eagles-Smith et al. 2008, 2009a, b) and are used only in correlation analyses with HOCs to assess patterns in bioaccumulation. Liver THg and Se concentrations were determined at the U.S. Geological Survey, Davis Field Station Mercury Lab (Davis, CA) and by the Trace Element Research Lab, Texas A&M University,

College Station, Texas, USA, respectively, following Eagles-Smith et al. (2009a).

P450 EROD Activity

Ethoxyresorufin-*O*-dealkylase activity was assayed in liver microsomes of all Forster's tern and Caspian tern samples collected during 2005 using the method of Burke and Mayer (1983) as adapted to a fluorescence microwell plate scanner (Melancon 1996). Hepatic microsomes were prepared from homogenates of thawed liver samples by differential centrifugation. The 11,000 *g* supernatant was centrifuged at 40,000 *g* for 60 min to obtain the microsomal pellet. Each 100,000 *g* pellet was resuspended in 4.0 mL/g tissue weight of 0.05 M Na/KPO₄ and 0.001 M disodium ethylenediamine tetraacetate, pH 7.6. The 260-mL total assay volume contained microsomes equivalent to 1.3 mg of liver, 1.25 μ M substrate, and 0.125 mM nicotinamide adenine dinucleotide phosphate-H in 0.066 M Tris-HCl buffer, pH 7.4. Protein concentrations were determined using the BCA Assay Kit (Pierce Chemical Company, Rockford, IL, USA) as adapted to a microwell plate scanner. EROD activity was calculated as picomoles of product formed per minute per milligram of microsomal protein.

Quality Assurance

Limits of detection for PCB and PBDE averaged 0.0001 μ g/g \pm 0.00001 SD, 0.003 μ g/g \pm 0.001 SD, and 0.006 μ g/g \pm 0.001 SD for organochlorine pesticides. Spiked recoveries for detected organic analytes averaged 95.6 \pm 20.5% SD. Procedural blanks contained either no or insignificant traces of organic compounds.

Data Analyses

We summed concentrations of all PBDEs, PCBs, and pesticides and report \sum PBDEs ($n = 38$ congeners),

Table 1 Halogenated organic compounds measured in livers of adult Caspian terns and Forster's tern adults and chicks in San Francisco Bay, California, 2005

HOCs	Members measured
Polybrominated diphenyl ethers	1, 2, 3, 7, 8/11, 10, 12, 13, 15, 17, 25, 28, 30, 32, 33, 35, 37, 47, 49, 66, 71, 75, 77, 85, 99, 100, 116, 118, 119, 126, 138, 153, 154, 155, 166, 181, 183, 190
Polychlorinated biphenyls	1, 7/9, 8/11, 10, 12, 13, 14, 15, 16, 17, 18, 19, 22/51, 24/47, 25, 26, 28, 29, 30, 31, 33/20, 39, 40, 41/64, 42/59, 44, 45, 46, 47/75, 48, 49, 52, 53, 60/56, 63, 66, 67, 69, 70, 72, 74/61, 77, 81, 82, 83, 84, 85, 87/115, 92, 95/80, 97, 99, 101/90, 105, 107, 110, 114, 118, 119, 126, 128, 129, 130, 135, 138/160, 141/179, 146, 149/123, 151, 153/132, 156, 157/173, 158, 166, 167, 169, 170/190, 171/202, 172, 174, 175, 176/137, 177, 178, 180, 183, 185, 187, 189, 191, 193, 194, 195/208, 196, 197, 199, 200, 205, 206, 207, 209
Dichloro-diphenyl-trichloroethane	<i>p,p'</i> -DDE, <i>o,p'</i> -DDD, <i>o,p'</i> -DDE, <i>o,p'</i> -DDT, <i>o,p'</i> -DDD, <i>p,p'</i> -DDT

\sum PCBs ($n = 101$ congeners), and \sum DDTs (6 compounds of DDE, DDD, DDT). PCBs were further analyzed based on the homologue grouping that accounted for >80% of all \sum PCBs (di-, tri-, penta-, hexa-, and hepta-chlorinated compounds) into their respective homologues based on preliminary profile charts. We did not separate dioxinlike PCBs (IUPAC Nos. 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189) in these analyses because in a preliminary step we found that their concentrations were highly correlated with \sum PCBs ($R^2 = 0.99$, $p < 0.0001$) and therefore we expected response in \sum PCBs to reflect the dioxinlike PCBs. Similarly, \sum PBDEs were further analyzed based on the two groups that accounted for >80% of all \sum PBDEs (tetra and penta) based on preliminary profile charts. All DDT, PCB, and PBDE concentrations are reported on a wet weight basis ($\mu\text{g/g}$) to be consistent with reported concentrations in the literature. Because we sampled both adult and chick Forster's terns, but only adult Caspian terns, we used a two-tiered approach to analyze HOC data, dependent upon the species and age. First, for adults only, we used multivariate analysis of covariance (MANCOVA; JMP 2001) to test for differences among species, region (North Bay, Central South Bay, and Lower South Bay), sex, and nesting stage for \sum PBDEs, \sum PCBs, and \sum DDTs. Next, we used MANCOVA to test for differences in the same response variables between regions and ages (adult and chick) for Forster's terns only. We used the same two analyses to test for differences in the concentrations of the PCB and PBDE homologues. For all analyses we included % liver lipid as a covariate to control for differences in \sum DDT, \sum PCB, and \sum PBDE concentrations associated with lipid content. When MANCOVA results were significant, we used analysis of covariance (ANCOVA; JMP 2001) to examine those differences. To assess relationships between HOC classes, we used Pearson-product moment correlation analysis among \sum PBDEs, \sum PCBs, and \sum DDTs for each species. To determine if HOCs were correlated with Hg and Se, we used Pearson-product moment correlation analysis. When sample concentrations were below detection limits, we inserted one-half of the detection limit value (Augsburger et al. 2008; Ricca et al. 2008). All data were \log_e -transformed prior to analyses and mean \log_e -concentrations are presented as geometric means \pm SE based on the back-transformed least squares mean \pm SE from the model output. The SEs of back-transformed values were estimated using the Delta method.

P450 EROD

We used a three-tiered approach to analyze P450 EROD data. First, using a larger dataset for which we had measured P450 EROD activity in all birds collected, we used a four-way analysis of variance (ANOVA) to determine if

P450 EROD activity differed by species, region, sex, and breeding stage (prebreeding or breeding) in adult Caspian terns and Forster's terns. Second, we used a three-way ANOVA to determine if P450 EROD activity differed by age (adult or chick), region, or nesting stage in Forster's terns. We included all two-way interactions of main effects and we removed interaction terms from the final model if they were not significant. Post hoc tests were conducted using Tukey's HSD to protect for Type I error. Finally, because of the limited dataset for which we measured both HOCs and P450 responses, we used Pearson-product moment correlation analysis to assess relationships between HOC classes and P450 stress measures for each species and age (adult and chick). This approach allowed us to determine if in fact P450 stress markers responded to differences in HOCs in the SFB.

Results

PBDEs

\sum PBDEs were dominated by tetra- and penta-BDEs. Tetra-BDE (principally BDE 47) and penta-BDEs (primarily BDE 99 and 100) composed 70%, 76%, and 51% of \sum PBDEs in adult Caspian terns, adult Forster's terns, and Forster's tern chicks; respectively. Moreover, tetra-BDE 47 constituted 42%, 53%, and 33% of \sum PBDEs found in adult Caspian terns, adult Forster's terns, and Forster's tern chicks; respectively, whereas penta-BDE 99 and 100 composed 28%, 23%, and 18% of \sum PBDEs in adult Caspian terns, adult Forster's terns, and Forster's tern chicks; respectively. All other individual PBDE congeners composed <3% of \sum PBDEs.

PCBs

\sum PCBs were dominated by five main homologue groups: di- (IUPAC 4–15), tri- (IUPAC 16–39), penta- (IUPAC 82–127), hexa- (IUPAC 128–169), and hepta- (IUPAC 170–193) chlorinated PCB homologues composed 95%, 93%, and 94% of \sum PCBs in adult Caspian terns, adult Forster's terns, and Forster's tern chicks; respectively (Figs. 1, 2). No other homologue group made up more than 3% of the \sum PCBs. Tri- (range = 38–39%), hexa- (range = 19–20%), and hepta- (range = 15–17%) chlorinated PCB homologues composed the majority of \sum PCBs, whereas di- (range = 9–13%) and penta- (range = 8–9%) chlorinated homologues were less common (Figs. 1, 2). Individual IUPAC congener 15, 16, 17, 138/160, 153/132, 180, and 187 together made up 68%, 69%, and 66% of \sum PCBs in adult Caspian terns, adult Forster's terns, and Forster's tern chicks; respectively (Figs. 1, 2). Of those individual

congeners, the lighter, less chlorinated congeners (IUPAC 15, range = 7–9%; IUPAC 16, range = 19–21%; and IUPAC 17 range 15–17%) together made up 43–45% of \sum PCBs, whereas the more chlorinated congeners that are known to bioaccumulate in marine and estuarine environments together made up 23–24% of \sum PCBs (IUPAC 138/160, range = 5%; IUPAC 153/132, range = 8–9%; IUPAC 180, range 6–9%; IUPAC 187, range = 3–4%; Figs. 1, 2). No other individual congeners made up more than 3% of the \sum PCBs. Dioxinlike PCB congeners (Poland and Glover 1977; Safe 1984) composed 3% of

each of the \sum PCBs in adult Caspian terns, adult Forster's terns, and Forster's tern chicks and mean \sum dioxinlike PCBs were 0.02 $\mu\text{g/g ww} \pm 0.01$ SE, 0.08 $\mu\text{g/g ww} \pm 0.03$ SE, and 0.03 $\mu\text{g/g ww} \pm 0.01$ SE; respectively.

DDTs

p,p'-DDE composed 98%, 90%, and 82% of all \sum DDTs detected in liver tissue samples from adult Caspian terns, adult Forster's terns, and Forster's tern chicks; respectively.

Fig. 1 PCB congener profiles in livers of Capian tern adults ($n = 5$) collected during the 2005 breeding season in the San Francisco Bay, California, USA. Mono, tetra, octa, and nona-deca homologue groups are not presented because they composed <5% of the total congeners

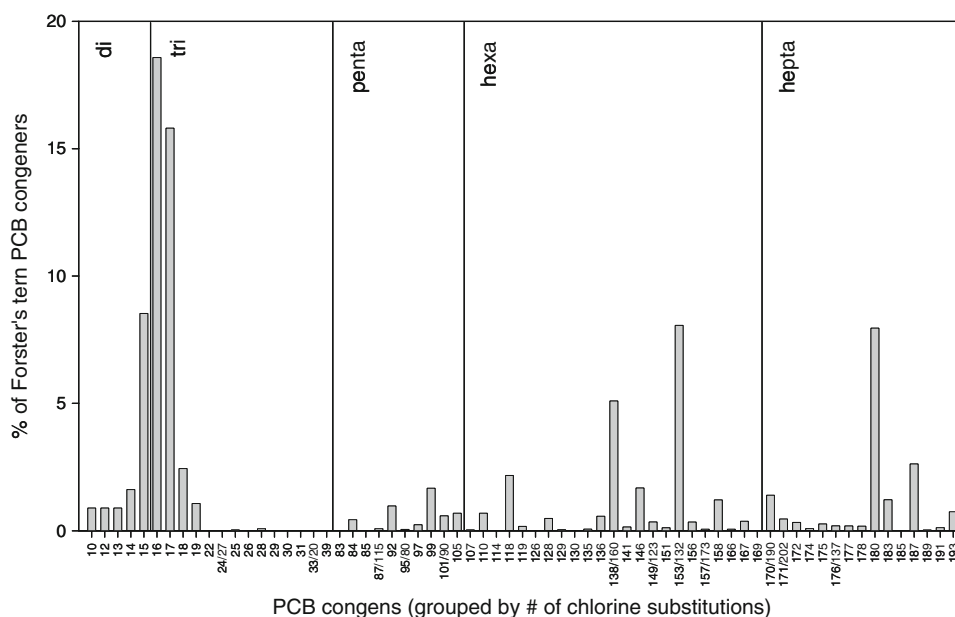


Fig. 2 PCB congener profiles in livers of Forster's tern adults and chicks ($n = 13$) collected during the 2005 breeding season in the San Francisco Bay, California, USA. Mono, tetra, octa, and nona-deca homologue groups are not presented because they composed <6% of the total congeners

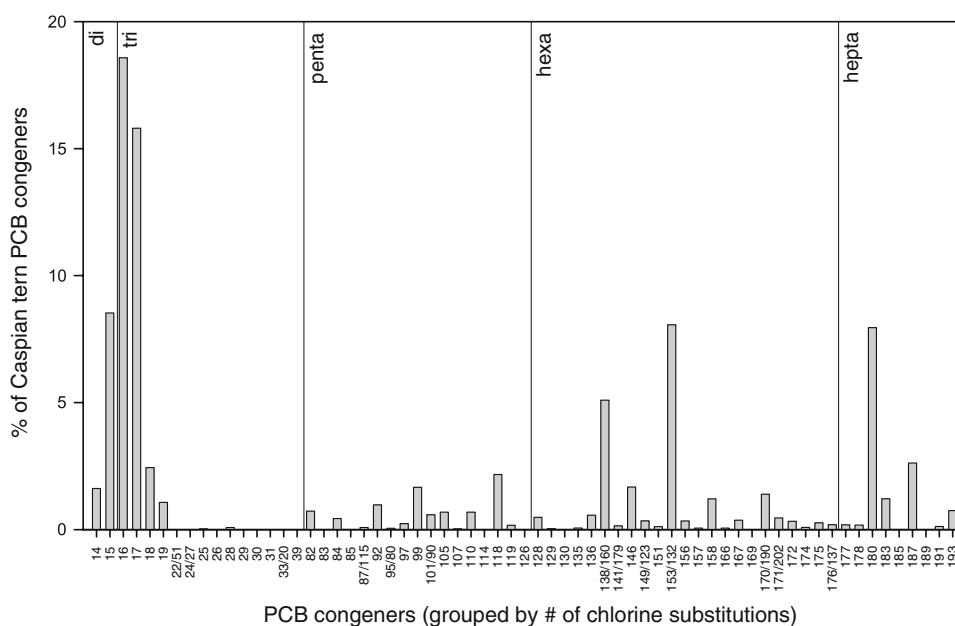


Table 2 Concentrations of Σ organochlorine compounds (PCBs, PBDEs, DDTs; $\mu\text{g/g ww}$) from livers of Caspian tern and Forster's tern adults and chicks in San Francisco Bay, California, 2005

Species	Region	Age	<i>N</i>	% Lipid ^a	SE ^b	ΣPCB^c	SE ^b	ΣPBDE^c	SE ^b	ΣDDT^c	SE ^b
Caspian tern ^{d,e}	North Bay	Adult	5	7.1	1.8	1.52	0.57	0.22	0.1	0.72	0.23
Forster's tern	North Bay	Adult	1	17.8		0.83		0.05		0.21	
Forster's tern	Central South Bay	Adult	2	15.7		11.31		0.64		0.62	
Forster's tern	Lower South Bay	Adult	4	12.1	2.4	4.08	2.24	0.24	0.13	0.41	0.19
Forster's ^f tern	Central South Bay	Chick	3	14.8	2.0	1.48	0.94	0.08	0.05	0.09	0.05
Forster's tern	Lower South Bay	Chick	3	15.0	3.2	1.09	0.69	0.04	0.02	0.07	0.04

^a % Lipid reported as arithmetic mean^b SE represents one standard error^c Σ concentrations of PCBs, PBDEs, and DDTs reported as geometric means^d No Caspian tern chicks were collected during study^e All Caspian terns were collected in the North Bay^f No Forster's tern chicks were collected in the North Bay

Comparisons Among Species, Regions, Sex, Breeding Stage, and Age

Concentrations of ΣPBDEs , ΣPCBs , and ΣDDTs were similar among species, regions, sexes, and breeding stage ($\lambda = 0.006$, $F_{18,8.97} = 2.59$, $p = 0.08$) for adult birds, although there was a nonsignificant trend of higher ΣPCBs and ΣPBDEs in adults from Central South Bay and Lower South Bay (Table 2) than North Bay. ΣPBDE ($F_{1,8} = 8.86$, $p = 0.01$), ΣPCB ($F_{1,8} = 9.99$, $p = 0.01$), and ΣDDT ($F_{1,8} = 7.19$, $p = 0.02$) concentrations were higher in adult Forster's terns than in Forster's tern chicks ($\lambda = 0.07$, $F_{12,16.16} = 2.30$, $p = 0.05$; Table 2). None of the PCB ($\lambda = 0.01$, $F_{30,6} = 0.39$, $p = 0.95$) or PBDE ($\lambda = 0.49$, $F_{12,8} = 0.27$, $p = 0.97$) homologues in adults differed by species, sexes, nesting stage or among regions nor did PCB ($\lambda = 0.13$, $F_{20,14.21} = 0.57$, $p = 0.87$) or PBDE homologues ($\lambda = 0.43$, $F_{8,14} = 0.90$, $p = 0.53$) differ between ages for Forster's terns.

Forster's tern ΣDDT was positively correlated with ΣPBDE and ΣPCB , and ΣPCB was positively correlated with ΣPBDE (Fig. 3). Similarly, Caspian tern ΣPCBs were positively correlated with ΣDDTs , but ΣPBDEs were not correlated with ΣDDTs or ΣPCBs (Fig. 3). ΣDDTs and liver tissue Se concentrations were positively correlated for Forster's terns and Caspian terns combined, and there was a nonsignificant positive trend between ΣPBDEs and ΣPCBs with Se (Fig. 4). Conversely, Forster's tern and Caspian tern ΣPBDEs , ΣPCBs , and ΣDDTs were not correlated with liver tissue THg (Fig. 4).

P450 EROD

The P450 EROD activity in adult Caspian terns and Forster's terns differed between species ($F_{1,47} = 39.63$,

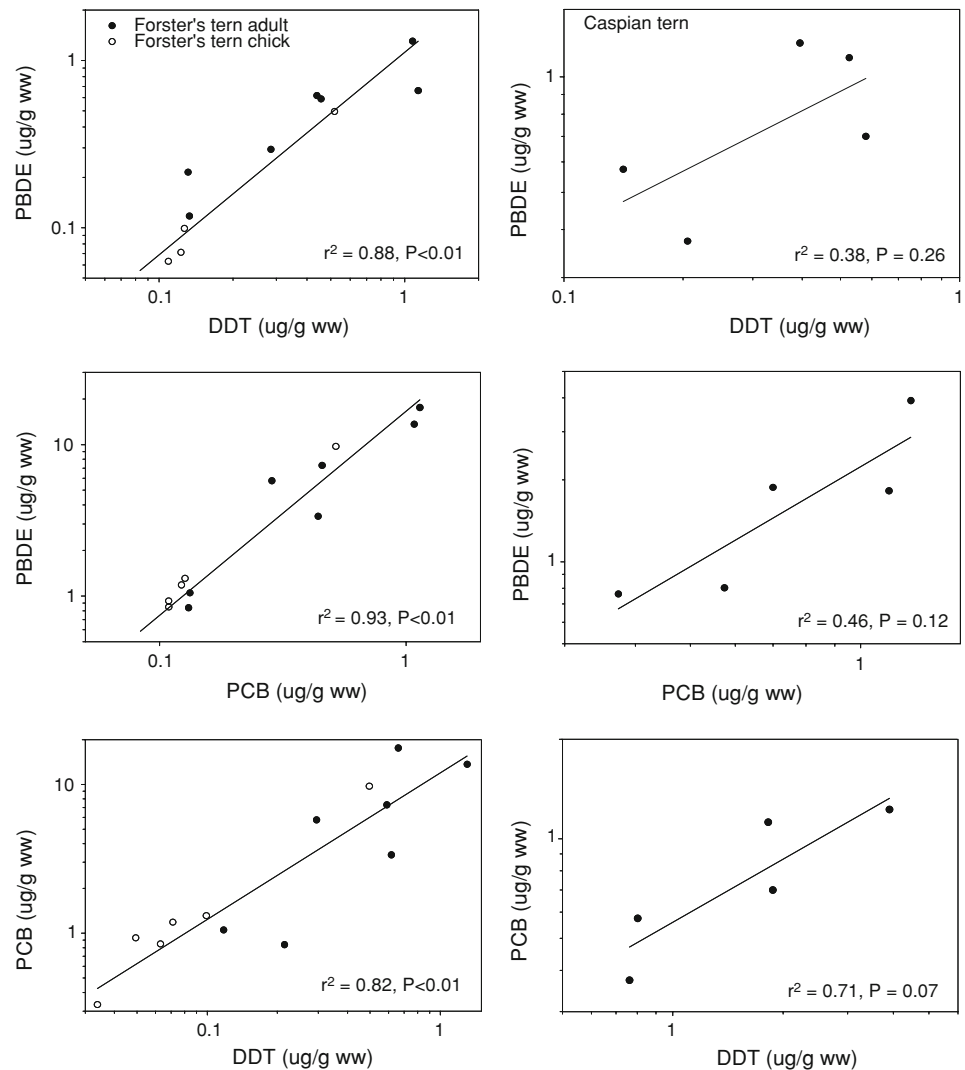
$p < 0.0001$) and nesting stage ($F_{1,47} = 5.48$, $p = 0.02$) but not among regions ($F_{2,46} = 0.04$, $p = 0.95$) or between sexes ($F_{1,46} = 0.15$, $p = 0.69$). P450 EROD activity was higher in adult Forster's terns than Caspian terns (Table 4). When analyzing only Forster's terns, we found a significant difference in P450 EROD activity between adults and chicks ($F_{1,50} = 30.20$, $p < 0.0001$) and among regions ($F_{2,50} = 5.22$, $p = 0.008$) but similar activity between nesting stages ($F_{1,50} = 1.48$, $p = 0.22$). P450 EROD activity was higher in Forster's tern adults than chicks; Forster's tern EROD activity was higher in the Central South Bay (Table 3). P450 EROD activity levels (mean \pm SE) are reported separately by species, age, region, and nesting stage for Caspian terns and Forster's terns in Table 3. P450 activity was not correlated with HOCs in either species, or age classes (Table 4).

Discussion

HOCs

The HOC contamination of waterbirds in SFB has been previously reported in several species, especially diving ducks (see Hothem et al. 1998; Miles and Ohlendorf 1993; Ohlendorf et al. 1991), but this is the first study within the last two decades to measure concentrations of HOCs in SFB waterbirds. Additionally, this is the first study specifically to assess HOC concentrations in Forster's tern and Caspian tern livers in the SFB. With a relatively limited sample size, we still observed spatial variation in contaminant concentrations for Forster's terns. Specifically, HOCs generally were higher at the Central South Bay and Lower South Bay sites, a result consistent with recent research on PCBs and PBDEs in Caspian tern and Forster's

Fig. 3 Correlation among Σ PBDEs, Σ PCBs, and Σ DDTs in livers of Caspian tern adults and Forster's tern adults and chicks



tern eggs (She et al. 2008) as well as with PBDEs in both water and sediment (Oros et al. 2005). Further, canvasbacks (*Aythya valisineria*; Hothem et al. 1998; Miles and Ohlendorf 1993), greater scaup (*A. marila*) and lesser scaup (*A. affinis*; Hothem et al. 1998), and surf scoters (*Melanitta perspicillata*; Ohlendorf et al. 1991) also had higher levels of PCBs in Lower South Bay compared to North Bay. Collectively, these results demonstrate that waterbird exposure to HOC contaminants is likely due to relatively distinct point-source inputs and generally follow the distribution patterns in water and sediment. It is unclear exactly why these trends occur, but they might be related to either historical land use, deposition from surrounding streams, or redistribution from other areas within the estuary.

Within the SFB, the Central South Bay and Lower South Bay differ from the northern portions of the bay in several aspects that might influence HOC availability. The Central

South Bay and Lower South Bay has a population base that is twice as large as the North Bay area, is a seasonally negative estuary (evaporation exceeds the freshwater supply from rivers and from local rain), receives less than 10% of the freshwater river runoff from the SFB area rivers but receives up to 76% of the total SFB wastewater, and has limited flushing capacity (Squire et al. 2002). Further, seasonal winds transport sediment from the North Bay to the Central South Bay and Lower South Bay (Lacy et al. 1996). Sediments within the SFB act as contaminant reservoirs and processes that result in potential accumulation of sediment in portions of the Bay might result in increased localized contaminant availability (Schoellhamer et al. 2007).

Potential sources of PCB and PBDE exposure might be inferred from the distribution of various congeners. For instance, the occurrence of more di- and tri-chlorinated PCB homologue groups is typically associated with

Fig. 4 Correlation among Σ PBDEs, Σ PCBs, Σ DDTs, and Hg and Se in livers of Caspian terns and Forster's tern adults and chicks

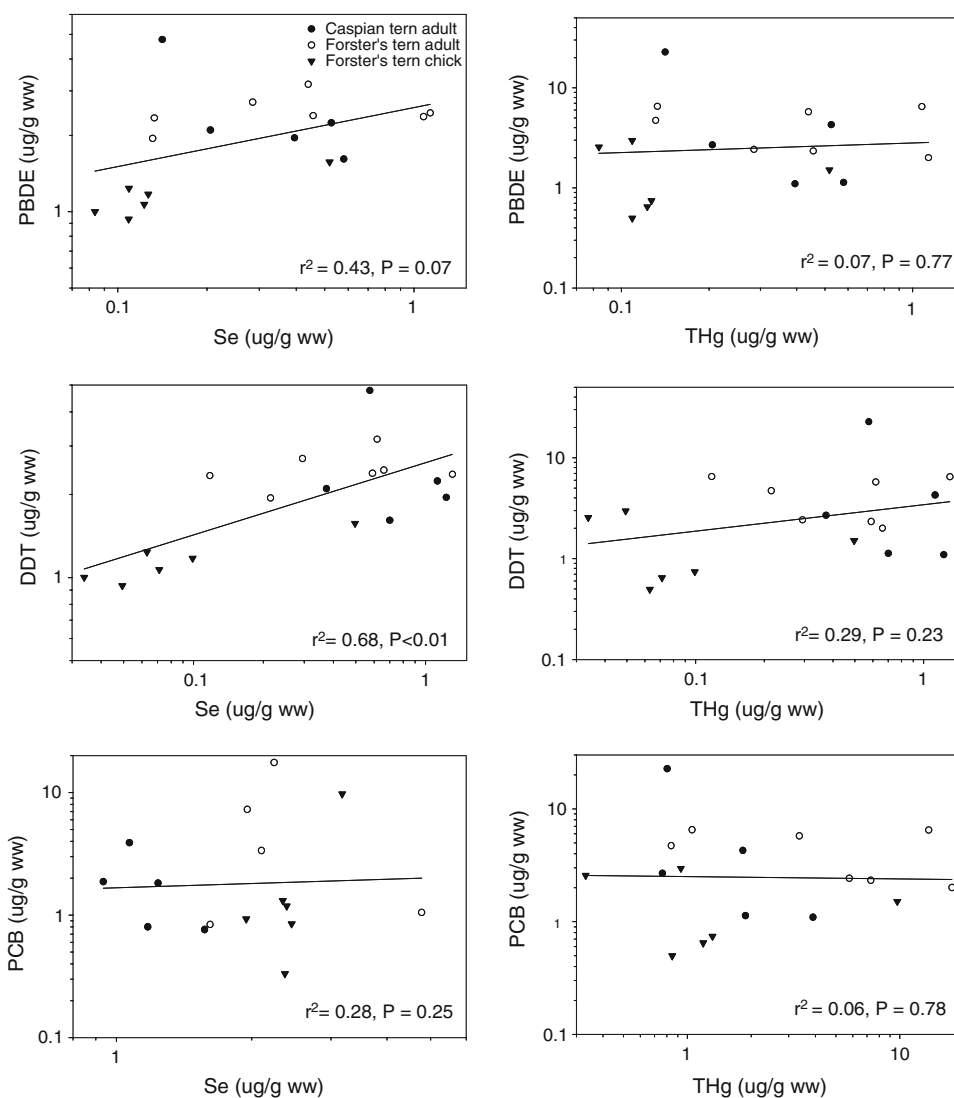


Table 3 Means (\pm SE) of P450 EROD activity (pmol/min/mg) in liver tissue samples from Caspian tern and Forster's tern adults and chicks in San Francisco Bay, California, 2005

Species and age	North Bay	Central South Bay	Lower South Bay	Prebreeding	Breeding
Caspian tern adult	2.5 \pm 0.12 (10)	—	—	2.5 \pm 0.12 (10)	—
Forster's tern adult	14.14 \pm 1.90 (21)	18.00 \pm 2.41 (4)	14.28 \pm 1.80 (21)	18.38 \pm 1.74 (18)	12.07 \pm 1.46 (28)
Forster's tern chick	—	12.0 \pm 2.77 (5)	0.72 \pm 0.27 (11)		

Note: Sample sizes are included in parenthesis

long-term decomposition of higher chlorinated PCBs (Blais 2005; Borga et al. 2001; Van den Brink 1997). Further, elevated concentrations of BDE 47, 99, and 100 are consistent typically with the penta-BDE commercial fire retardant mixture (Nordstrom et al. 2000; She et al. 2008). In this study we observed both increased di- and tri-chlorinated PCB homologue groups and elevated concentrations of BDE 47, 99, and 100. PCB results reflected the long-term legacy of PCB contamination in the SFB [see

review in Davis et al. (2007)], where the pattern of high proportions of di- and tri-chlorinated homologue groups were most likely a result of decomposition of higher chlorinated PCBs in the SFB. The pattern of elevated penta-BDEs is consistent with highly populated, urbanized sites, although the pathway by which PBDEs get into the environment is not fully understood (Alcock et al. 2003; Oram et al. 2008). Elevated concentrations of penta-BDEs also have been recently measured in Caspian

Table 4 Linear regression of P450 EROD activity on concentrations of \sum PBDEs, \sum DDTs, \sum PCBs from livers of Caspian tern (CATE) and Forster's tern (FOTE) adults and chicks in San Francisco Bay, California, 2005

Species	Age	HOC group	R^2	N	p	Regression coefficient (b)
CATE	Adult	\sum PBDE	0.03	5	0.76	−0.13
CATE	Adult	\sum DDT	0.00	5	0.38	−0.63
CATE	Adult	\sum PCB	0.00	5	0.92	−0.03
FOTE	Adult	\sum PBDE	0.12	7	0.42	−2.96
FOTE	Adult	\sum DDT	0.33	7	0.17	−7.42
FOTE	Adult	\sum PCB	0.14	7	0.40	−0.61
FOTE	Chick	\sum PBDE	0.00	5	0.98	0.06
FOTE	Chick	\sum DDT	0.04	6	0.69	−1.92
FOTE	Chick	\sum PCB	0.00	6	0.97	0.04

Note: In determining significant correlations we used the Bonferroni correction factor to control for site and collection period. Significance was determined for Caspian terns if $p \leq 0.05$, for Forster's tern adults if $p \leq 0.05/5$ (three sites and two collection periods), and for Forster's chicks when $p \leq 0.05/2$ (two sites)

terns and Forster's tern eggs (She et al. 2008) and fish tissue (Brown et al. 2006) in the SFB. In fact, She et al. (2008) reported one of the highest PBDE concentrations ever recorded in the world for wildlife in SFB Forster's tern eggs.

Concentrations of PCBs and DDT in SFB Forster's terns and Caspian terns were all lower than those found to be significantly harmful to adult birds. For instance, mallards (*Anas platyrhynchos*) fed a diet dosed with 25 $\mu\text{g/g}$ /day dry weight (dw) PCBs, resulting in mean liver PCB concentrations of 55.3 $\mu\text{g/g}$ dw (female) and 64.2 $\mu\text{g/g}$ dw (male), did not experience reproductive failure or mortality (Custer and Heinz 1980). The highest individual PCB concentration we observed in a SFB bird was 17.6 $\mu\text{g/g}$ ww, with the highest mean concentration of 4.1 $\mu\text{g/g}$ ww \pm 2.2 SE. Similarly, American black ducks (*A. rubripes*) fed 10 $\mu\text{g/g}$ /day dw DDE, resulting in mean carcass DDE concentrations of 151.5 $\mu\text{g/g}$ dw (male) and 159.6 $\mu\text{g/g}$ dw (female), had impaired reproduction, but no mortality occurred (Longcore and Stendell 1977). In our study, the highest DDT concentration we observed was 1.3 $\mu\text{g/g}$ ww, with a mean concentration of 0.5 $\mu\text{g/g}$ ww \pm 0.1 SE. Concentrations of DDTs do not appear to be high enough to result in impaired reproduction. Fernie et al. (2005) dosed American kestrel (*Falco sparverius*) chicks with 15.6 $\mu\text{g/g}$ /day ww of PBDE, resulting in carcass homogenate concentrations of 86.1 $\mu\text{g/g}$ ww, and observed increased measures of oxidative stress. In our study, the highest PBDE concentration we observed was 1.1 $\mu\text{g/g}$ ww, with a mean concentration of 0.3 $\mu\text{g/g}$ ww \pm 0.1 SE.

P450 EROD Activity

The EROD activity responses occurred in a predictable manner based on regional contaminant levels and foraging habitat exposure for Caspian terns and Forster's terns: higher contaminant concentrations in Central South Bay than in North Bay and higher in Forster's terns than in Caspian terns. Increased EROD activity in Forster's terns collected in the Central South Bay might be related to the general trend of higher HOCs (\sum PCBs and \sum PBDEs) that we observed in tern tissue for this region. Consistent with this finding, She et al. (2008) reported higher PCBs and PBDEs in Caspian tern and Forster's tern eggs in this area, and Oros et al. (2005) reported higher PBDEs in both water and sediment in this area. Further, She et al. (2008) also found that PCBs were higher in Forster's terns than in Caspian terns, similar to our results for the liver tissue samples in Forster's terns and Caspian terns.

Differences in EROD activity between species might be related to their differential use of foraging habitats and associated HOC exposure. Caspian terns tend to forage in deep, open water (Anderson et al. 2007; Lyons et al. 2005), whereas Forster's terns tend to forage in shallow water closer to shorelines and in wetlands (Ackerman et al. 2008a). Concentrations of PCBs tend to be higher in wetland habitats along the shorelines in the SFB (Davis et al. 2007). Further, differences between adult Forster's terns and chicks might be an effect of exposure time, with breeding adults having prolonged periods of exposure to HOCs in the SFB, whereas chicks had a shorter exposure period. The fact that we did not find a correlation between P450 EROD activity and HOC groups suggests that the relationship might not be linear; alternatively, the small sample sizes in our study might have precluded finding a correlation after controlling for species, region, and age differences. Further studies are warranted to elucidate the response of P450 EROD activity and HOCs in the SFB.

Summary

Current loading rates of \sum PCBs (CRWQCB 2004; Davis et al. 2007) and \sum PBDEs (North 2004; Oram et al. 2008) from sources into the SFB suggest that \sum PBDEs might be an order of magnitude greater than those of PCBs in source inputs and are not expected to decline in the near future based on projected estimates (Oram et al. 2008; Oros et al. 2005). If this is the case, then \sum PBDEs concentrations in Caspian terns and Forster's terns might be expected to increase until there is a reduction in source loadings. The effect of these high HOC concentrations on waterbird health and reproduction are unknown. Given the probable influence of other SFB wetland contaminants (e.g., Hg) to

impair waterbird egg and chick survival (Ackerman et al. 2007, 2008a, b) and similar bioaccumulation rates between HOCs and Se (this study), there might be interactions among contaminants in SFB (see Pollock and Machin 2008), which could produce possible antagonistic or ameliorative effects. Future research should investigate potential physiological and reproductive effects of HOCs with other SFB contaminants on waterbirds in SFB.

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